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Surface Effect on Optical Bistability in Coupled Exciton-Phonon Systems Inside a Cavity

by

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Surface Effect on Optical Bistability in Coupled  
Exciton-Phonon Systems Inside a Cavity

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Abstract

When a laser beam is directed on an exciton-phonon coupling system inside an optical cavity, the cavity field intensity is investigated as a function of the driving field intensity for various distances between the sample and a metallic surface representing one of the mirrors of the cavity. Optical bistability is found to be induced by the surface when the cavity field is on resonance with the excitonic transition frequency. It is also found that at the threshold, the direction of switch of the optical bistability can be controlled by adjusting the distance from the surface.

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The investigation of optical properties of molecular systems near a solid surface has been of great interest from both the fundamental and practical points of view. The study of an atom or a molecule adsorbed on a solid surface has attracted much attention<sup>1-4</sup> in the past, both theoretically and experimentally. Exciton-phonon coupling systems such as organic semiconductors have been considered<sup>5</sup> very recently instead of single molecular systems located near an ideal metal surface. Surface-induced optical bistability has been discovered and its nature and origin investigated.

In Ref. 5 we considered, as an example, a linear chain of polydiacetylene-toluene-sulfonate (PTS) oriented parallel to the metal surface. The system closely resembles the experimental situation<sup>6</sup> in which a single crystal thin film of polymer is formed on the solid substrate. Polydiacetylenes are important materials for study in view of a variety of applications in optical signal processing such as ultrafast all-optical switches and logic gates. One can foresee potential applications in ultrahigh-speed ( $\sim \text{TH}_z$ ) multiplexing-demultiplexing operation for telecommunication devices, because the ultrafast response (sub-picoseconds) and large nonlinearity make these materials ideal candidates.

On the other hand, we have also found novel optical bistability mediated by the exciton-phonon interaction in polymers inside an optical cavity.<sup>7</sup> It is remarkable that the hysteresis loop may follow the normal or reversed cycle depending on the frequency of the incident light. We now consider in this paper a PTS chain inside an optical cavity. The chain is embedded in a non-absorbing material of dielectric constant  $\epsilon_1$  and is oriented parallel to the reflecting metallic mirrors that constitute the cavity. The dielectric function of the mirror is

$$\epsilon_2(\omega) = 1 - \omega_p^2 / \omega(\omega + i\Gamma_p) \quad , \quad (1)$$

where  $\omega_p$  is the plasma frequency and  $\Gamma_p$  the damping rate. Our purpose is to investigate how the optical bistability behavior is influenced by the presence of the metallic surface of one of the mirrors if the polymer is placed at a distance  $d$  smaller than the excitonic wavelength. When an incident laser beam is directed into the cavity, excitons are excited in PTS and act as the emitting dipoles. The emitted light is reflected by the surface, and the reflected light interacts in turn with the emitting excitons and changes their response to the incident light. At the same time, the excitons couple with phonon modes in PTS, which play the role of mediation in the photon-exciton interaction.<sup>8</sup>

To make the complicated interacting system manageable, we model the excitons, phonon modes and cavity field as damped oscillators and assume a dissipative interaction between the exciton and surface-reflected field.<sup>3</sup> The problem is then treated by means of Dekker's quantization procedure for dissipative systems.<sup>9</sup> In this model, the system is described by the non-Hermitian Hamiltonian

$$H = (\Omega - i\kappa)a^\dagger a + (\omega_x - i\gamma_x)c^\dagger c + \sum_i (\omega_i - i\gamma_i)b_i^\dagger b_i + \sum_i \lambda_i c^\dagger c (b_i^\dagger + b_i) \\ + ig (a^\dagger c - c^\dagger a) + i(a^\dagger E_o e^{-i\omega_o t} - a E_o^* e^{i\omega_o t}) - \mu^* a^\dagger E_R \quad , \quad (2)$$

where  $a^\dagger$  ( $a$ ),  $b_i^\dagger$  ( $b_i$ ) and  $c^\dagger$  ( $c$ ) stand for the creation (annihilation) operators for the cavity field, the  $i$ -th phonon mode and the exciton with corresponding frequencies  $\Omega$ ,  $\omega_i$  and  $\omega_x$ , respectively.  $\kappa$ ,  $\gamma_x$  and  $\gamma_i$  are the

damping rates for the cavity field, exciton and  $i$ -th phonon modes, respectively.  $\lambda_i$  is the coupling constant for the  $i$ -th phonon modes interacting with the exciton, and  $g$  is the exciton-cavity field coupling constant. The frequency and amplitude of the coherent driving field are denoted by  $\omega_0$  and  $E_0$ , respectively. The dipole moment of the emitting exciton is represented by  $p = \mu a$  with  $\mu$  as its matrix element.  $E_R$  stands for the reflected field at the position of the dipole; it is proportional to  $p$  and hence can be written as  $E_R = E_r a$ , where  $E_r$  is a c-number. As we shall see later,  $E_r$  is in general a complex quantity, so that the interaction is dissipative.

With the dissipative system characterized by the canonical variables  $a^\dagger$ ,  $a$ ,  $b_i^\dagger$ ,  $b_i$ ,  $c^\dagger$  and  $c$ , we have according to Dekker<sup>9</sup> the quantum Liouville equation

$$\begin{aligned} \dot{\rho} = & -i[a^\dagger, [a, H]\rho] + i[\rho[H^\dagger, a^\dagger], a] - i \sum_i [b_i^\dagger, [b_i, H]\rho] \\ & + i \sum_i [\rho[H^\dagger, b_i^\dagger], b_i] - i[c^\dagger, [c, H]\rho] + i[\rho[H^\dagger, c^\dagger], c] \quad (3) \end{aligned}$$

Since we are mainly interested in possible optical multistability in our study of the relation between intensities of the cavity field and input field, we may take a semiclassical approach by neglecting all quantum fluctuations.<sup>10</sup> Thus, instead of the operators  $a$ ,  $b_i$ ,  $c$  and  $c^\dagger c$ , we are dealing with their mean values  $\alpha = \langle a \rangle$ ,  $\beta_i = \langle b_i \rangle$ ,  $\eta = \langle c \rangle$  and  $n = \langle c^\dagger c \rangle$ . With these mean values defined as classical variables, the corresponding equations of motion can be obtained from Eq. (3) as follows:

$$\dot{\alpha} = -(i\Delta_1 + \kappa)\alpha + g\eta + E_0, \quad (4a)$$

$$\dot{\beta}_i = -(i\omega_i + \gamma_i)\beta_i - i\lambda_i n, \quad (4b)$$

$$\dot{\eta} = -(i\Delta_2 + \gamma)\eta - i \sum_i \lambda_i (\beta_i + \beta_i^*)\eta - g\alpha, \quad (4c)$$

$$\dot{n} = -g(\alpha\eta^* + \alpha^*\eta) - 2\gamma n, \quad (4d)$$

where we have defined the detunings  $\Delta_1 = \Omega - \omega_0$  and  $\Delta_2 = \omega_x - \omega_0 + \omega_s$ ,  $\gamma = \gamma_x + \gamma_s$ . The surface effect on the exciton is reflected in the surface-induced frequency shift

$$\omega_s = |\mu|^2 \text{Re}(E_r/\mu) \quad (5a)$$

and the decay rate

$$\gamma_s = |\mu|^2 \text{Im}(E_r/\mu). \quad (5b)$$

The reflected electric field  $E_r$  in the dipole direction can be found by a classical approach. We take the oscillating dipole moment to be located at a distance  $d$  from the surface of a semi-infinite metal. From electromagnetic field theory, we find in a straightforward manner that

$$E_r = -i\sqrt{\epsilon_1} k_0^3 \mu \int_0^\infty du u^3 R_{\parallel} \exp(2i\mu_1 x)/\sqrt{1-u^2}. \quad (6)$$

In the derivation of Eq. (6), we have assumed that the dipole is perpendicular to the surface. Here we have also defined  $k_0 = \omega_x/c$ ,  $x = \sqrt{\epsilon_1} k_0 d$  and

$$R_{\parallel} = \frac{\epsilon_1 \mu_2 - \epsilon_2 \mu_1}{\epsilon_1 \mu_2 + \epsilon_2 \mu_1}, \quad (7a)$$

$$\mu_l = \sqrt{\epsilon_2/\epsilon_1 - \mu^2}, \quad l = 1, 2. \quad (7b)$$

Substituting Eq. (6) in Eqs. (5), we find

$$\omega_s = \frac{3}{2} \text{Im} \int_0^{\infty} du u^3 R_{\parallel} \exp(2i\mu_1 x)/\mu_1 \quad (8a)$$

$$\gamma_s = -\frac{3}{2} \text{Re} \int_0^{\infty} du u^3 R_{\parallel} \exp(2i\mu_1 x)/\mu_1, \quad (8b)$$

which have been expressed in the unit of  $\gamma_x = \frac{2}{3} \sqrt{\epsilon_1} |\mu|^2 \omega_x^3/c^3$ . From now on, and throughout this paper, all energies will be measured by this unit. If we introduce a dimensionless input field intensity  $I_{in} = g^2 |E_0|^2 / (\kappa \gamma_x)^2$ , we find from Eqs. (4), after some algebraic manipulation, that the mean exciton number  $n$  satisfies the cubic equation

$$\begin{aligned} & \lambda_p^2 [1 + (\Delta_1/\kappa)^2] n^3 + 2\lambda_p [\Delta_1 (g^2 - \Delta_1 \Delta_2)/\kappa^2 - \Delta_2] n^2 \\ & + \{ [1 + \gamma_s + (g^2 - \Delta_1 \Delta_2)/\kappa]^2 + [\Delta_2 + (1 + \gamma_s) \Delta_1/\kappa]^2 \} n - I_{in} = 0. \end{aligned} \quad (9)$$

At the same time, the cavity field intensity  $I_{cav} = |\alpha|^2/\gamma_x^2$  can be expressed in terms of the input field intensity  $I_{in}$  as



$$I_{\text{cav}} = \frac{(1+\gamma_s)^2 + (\Delta_2 - \lambda_p n)^2}{[1+\gamma_s + g^2/\kappa - \Delta_1(\Delta_2 - \lambda_p n)/\kappa]^2 + [\Delta_1(1+\gamma_s)/\kappa + \Delta_2 - \lambda_p n]^2} I_{\text{in}} \quad (10)$$

where we have defined  $\lambda_p = 2 \sum_i \lambda_i^2 / \omega_i$ . In the limit  $\kappa \rightarrow \infty$ , we are dealing with a bad cavity, namely, the cavity field damps out almost immediately. Equations (9) and (10) then reduce to

$$\lambda_p^2 n^3 - 2\lambda_p \Delta_2 n^2 + [(1+\gamma_s)^2 + \Delta_2^2] n - I_{\text{in}} = 0 \quad (11)$$

$$I_{\text{cav}} = I_{\text{in}} \quad (12)$$

which are identical to the results of Ref. 5, as expected.

Equation (9) is solved numerically, and the solutions are used to study the tristability of the cavity field intensity in Eq. (10) under various conditions. When the incident field frequency is on resonance with both the cavity field and the excitonic transition, the occurrence of optical bistability depends on the location of the chain in the cavity. If the chain is far away from the mirror surfaces of the cavity, Eq. (9) has only one physical solution and there is no optical bistability. If the PTS chain is near a mirror surface or if  $d$  is smaller than the excitonic wavelength, Eq. (9) may possess three real solutions, and it is possible to find optical bistability in the cavity. As discussed in Ref. 5, this surface-induced bistability is caused by both the frequency shift  $\omega_s$  and the width change  $\gamma_s$  of the excitonic transition due to the surface. If the distance becomes too small or  $x \ll 1$ , the surface absorption becomes so strong that the lifetime of

the exciton is too short to interact strongly and nonlinearly with the phonon modes. Consequently, there can no longer be bistability.

Off resonance, the behavior depends very much on the sign of the detuning. When  $\Delta_1 \geq 0$ , the optical bistability is expressed in a normal hysteresis loop,<sup>5</sup> and the presence of a surface can not change this nature. However, a nearby surface can affect the threshold input intensity and the contrast of the hysteresis loop. When  $\Delta_1 < 0$ , optical bistability can usually be found with the inverted hysteresis loop, even if the sample is far away from the surface. In the presence of a nearby surface, our calculations show a completely new phenomenon, namely that the two cavity field intensities cross each other as the driving field intensity changes.

In our numerical work, we have included all four phonon-modes that couple most strongly to the excitons.<sup>7,8,11</sup> These modes are characterized by the parameters  $\omega_1 = 5.16$ ,  $\lambda_1 = 2$ ;  $\omega_2 = 3.68$ ,  $\lambda_2 = 1.66$ ;  $\omega_3 = 2.98$ ,  $\lambda_3 = 0.46$ ;  $\omega_4 = 2.36$ ,  $\lambda_4 = 0.48$ . Some of the results for  $\Delta_1 < 0$  are plotted in Fig. 1 in which we demonstrate the change of the bistability nature by varying the distance from the surface only. Figure 1(a) shows no bistability because of the small distance. As mentioned above, the strong absorption by the surface prevents the exciton from interacting with phonon modes nonlinearly. There can not be optical bistability for any  $\Delta_1$ . In Figs. 1(b) and 1(c), we find the interesting new phenomenon where the two output states cross each other, forming a peculiar 8-shaped loop. The cavity field changes the normal bistability as the driving field intensity increases, but follows the inverted loop as  $I_{in}$  decreases. The hysteresis loop becomes a triangle in Fig. 1(d) at the distance  $x = 2.0$ . After this point, the loop becomes inverted and stays that way as the distance increases further.

As discussed in Ref. 7 in more detail, in an optical cavity, the real part of the optical response of PTS or the dispersion part plays a more important role than the absorption part because the refractive index is changed by the excitons which are excited by the input field. Our numerical computations indicate that the major contribution to the bistability comes from the exciton energy shift induced by the surface. This is in contrast to the case discussed in Ref. 5, where the absorptive bistability is caused by reduced vacuum fluctuations.

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**Figure Captions**

1. Cavity field intensity  $I_{\text{cav}}$  versus the incident field intensity  $I_{\text{in}}$  for various distances  $x$  between this system and the metal surface when  $\Delta_1 = -0.25$ ,  $\omega_x/\omega_p = 0.8$ ,  $\Delta_2 = 6$ ,  $g^2/k = 0.5$ . (a)  $x = 0.5$ , (b)  $x = 0.75$ , (c)  $x = 1.25$ , (d)  $x = 2.0$ , (e)  $x = 2.5$ , (f)  $x = \infty$ .

Fig. 1

